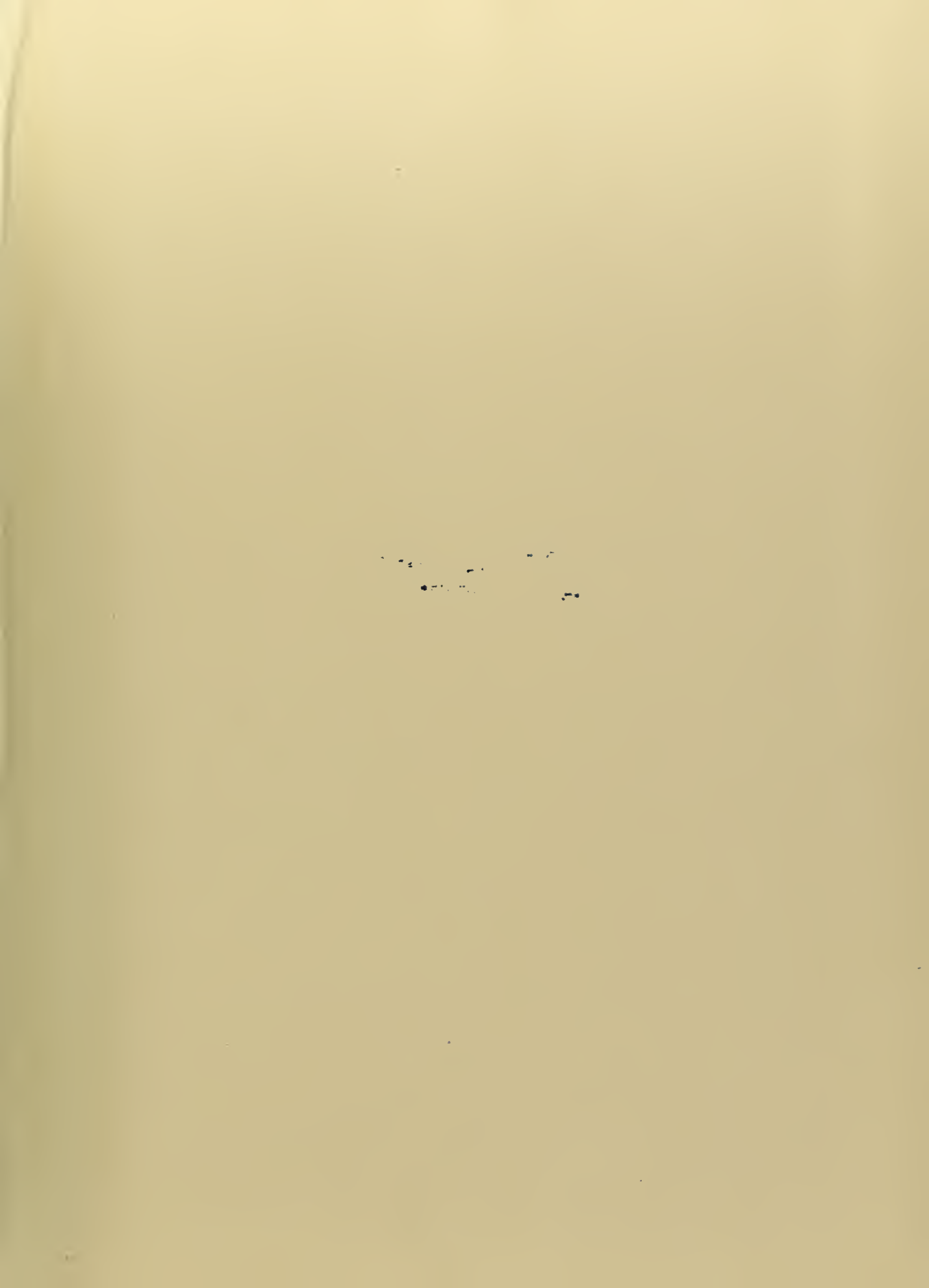


EQUIPMENT FOR THE PRODUCTION AND
STUDY OF CRYSTAL IMPERFECTIONS

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EQUIPMENT FOR THE PRODUCTION
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* * * *

Walter W. Fade

and

John A. Bryan



EQUIPMENT FOR THE PRODUCTION
AND
STUDY OF CRYSTAL IMPERFECTIONS

by

Walter Wayne Fade

Major, United States Army

and

John Andrew Bryan

Captain, United States Army

Submitted in partial fulfillment
of the requirements
for the degree of

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IN
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1955

Thesis

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This work is accepted as fulfilling
the thesis requirements for the degree of

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from the

United States Naval Postgraduate School



PREFACE

With current developments in the industrial and scientific worlds, everyday more stringent requirements must be met by our limited number of metals. It is with this in mind that we endeavor to learn more about the metals we have. Recent advancements in the field of cryogenics has afforded an excellent tool for the study of metals at low temperatures and metal crystal lattice theory. Although the authors contributed little in the way of experimental data, it is felt that the equipment built can contribute a great deal of valuable information in the field of crystal imperfections. It is hoped that the potentialities will be realized and this work will be continued.

The authors would like to express their appreciation to Professor E. C. Crittenden, Jr. for his guidance and council.

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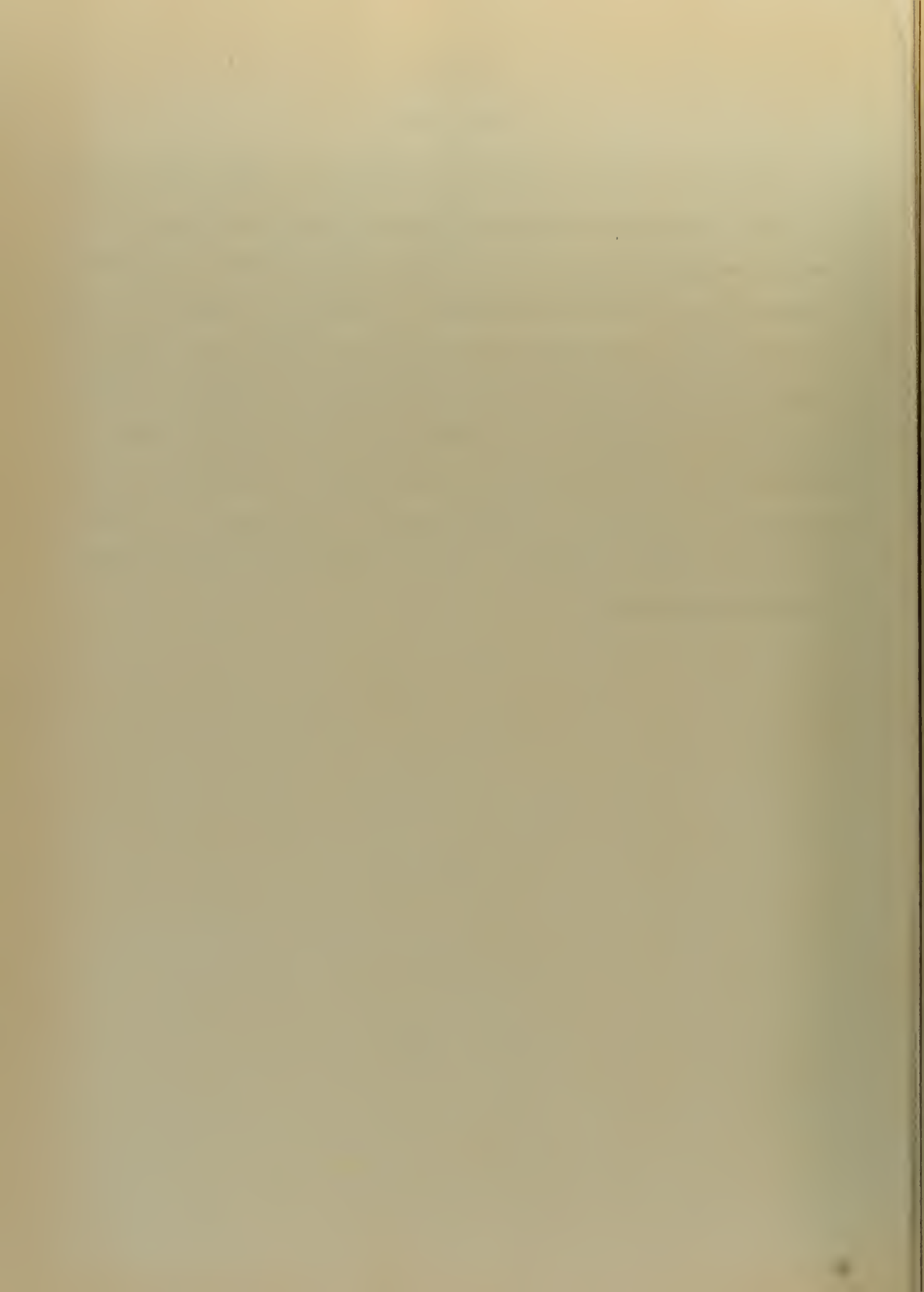
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CHAPTER I

INTRODUCTION

Within the overlapping fields of metallurgy and solid state physics, it is generally agreed that crystal lattice imperfections play a vital role in the physical properties of metals. Conventionally there are six primary types of crystal imperfections, 1 namely, (a) phonons, (b) electrons and holes, (c) excitons, (d) vacant lattice sites and interstitial atoms, (e) foreign atoms in either interstitial or substitutional positions and (f) dislocations. At present, the types of imperfections which appear to have the greatest effect on physical properties are vacancies and interstitials, foreign atoms, and dislocations. The units designed and herein described are for the production and study of these types of imperfections.



CHAPTER II

METAL FILM EVAPORATING UNIT

The development of a metal film evaporator unit for the production and pursuant study of known types of imperfections in metal crystals was undertaken with the primary intent of eliminating those physical experimental difficulties which have lead to question of the work by Crittenden and Hoffman, [2], Anders, [3], and Daniels [4] on thin films.

Primarily, the basic reason for metal films having acquired such a bad reputation as specimens of bulk metals, is that pressures ordinarily used for evaporation lead to a relatively high degree of contamination by the residual gasses. For example, at a pressure of 10^{-5} mm of mercury, each surface atom of a sample is struck approximately twice each second by a residual gas molecule. It is obvious, that under such a bombardment, some inclusion of foreign atoms within the crystal lattice of the evaporated metal, in either substitutional or interstitial positions, will occur. Such inclusions were the bane of previous investigators for they not only materially affected the properties exhibited by the metal film as distinguished from those exhibited by bulk metal, grown from melt, but were a type of imperfection which could not be reproduced from specimen to specimen.

By eliminating the inclusion of foreign atoms in the metal film, it will be possible, with this unit, to study several other facets of solid state physics in addition to the study of crystal imperfections.

Basically, the film evaporating unit (see Figure 1) was developed utilizing the forepump, parallel diffusion pumps and base plate from a



standard U.S. Navy MK 5 Optical Filming unit. The 18 inch diameter glass dome ordinarily used as part of the unit was cut off to form an open-ended cylinder 18 inches high. On this was mounted a removable 3/4 inch thick aluminum top plate. This arrangement provided two surfaces, the base plate and the top plate, through which electrical connections and insertions could be made into the central chamber.

In a test of the equipment of that portion of the unit below the glass cylinder a vacuum 2×10^{-5} mm of mercury was attained (see Figure 2).

Radially installed near the bottom, and within the glass cylinder, is a brass pan filled with activated charcoal (see Figure 3). This brass "charcoal pump" is so constructed as to have an externally controlled heating unit and a liquid nitrogen quenching tube built into the pump housing. During the pumping stage and until shortly before the evaporating cycle, the charcoal within the pump is heated and thus degassed. Upon attaining a vacuum of the order of 10^{-6} mm of mercury (Hg) within the heated cylinder the charcoal is quenched by cooling with liquid nitrogen. The charcoal then exhibits a strong affinity for absorbing and holding gas molecules. The volume of charcoal supplied is such that for 180 cubic inches of space within the sealed cylinder one cubic inch of charcoal is supplied.

Suspended from the top plate of the unit is a thermal shield (see Figure 3), consisting of a 16 inch diameter copper drum of 1/8 inch metal which has been wound, top and sides, with 5/8 inch copper tubing and with the lower end open. This thermal shield serves two purposes in providing the desired atmosphere: one, it can be heated to any desired temperature with either hot water or steam during the initial pumping



operations, thus, in effect, acting as a degassing unit; and two, just prior to and during the evaporation cycle, it can be cooled to -196°C by pouring liquid nitrogen through the coils, thus affording a condensing surface for the water vapor, oil vapor and carbon dioxide molecules remaining within the central chamber.

Another feature constructed within the evacuated cylinder is a liquid nitrogen cooled dual evaporation furnace (see Figure 3). This furnace consists of a 4 inch diameter brass pot wound with a $5/8$ inch copper tube. The pot is divided into two equal compartments, each with an externally controlled electrical circuit whereby either of the furnaces may be independently operated without heating the other. This feature allows the operator to evaporate a metal sample onto a substrate and then plate the entire surface of the specimen with a protective agent, such as quartz, while under an initial vacuum. This operation stabilizes the specimen against chance phenomena such as corrosion or oxidation while continuing the tests of the metal film.

All of the above features which require the use of liquid nitrogen are provided with a special entrance seal (see Figure 4) which is constructed in such a way that each copper tube connection is insulated from the brass sealing plug, and thus from the rest of the unit, by 4 inches of .020 stainless steel tubing. Utilizing the lower conductivity of stainless steel as an insulating factor, materially reduces the volume of metal which must be cooled to achieve the internal temperatures desired, thus providing better control within the evacuated cylinder.

Initially it was planned to construct the top plate so that two basic types of substrates could be utilized.



The simplest system developed was to utilize the bottom of a glass beaker as a substrate. The beaker employed has a slightly flared top, is $2\frac{1}{2}$ inches in diameter, and 6 inches in depth and has no pouring spout. An opening through the top plate, large enough to receive the body of the beaker but not the flared lip, is equipped with a rubber gasket and a ring to clamp the beaker down tightly. This arrangement places the bottom of the beaker into the cavity to be evacuated but leaves the mouth open to the atmosphere. Temperature control of the substrate is attained by the addition of coolants directly into the beaker. This system does not provide a means for annealing the metal film and therefore, is generally limited to studies involving the activation energy of metals and statistical studies of crystal nucleation.

A more complex holder (see Figure 5) was designed to be utilized with flat strips of either mica or fire polished glass as substrates with liquid helium as a coolant.

The device consists of a coolant chamber, a steel thermal web, a specimen holder, a filling tube $1/2$ inch in diameter of .006 inch low conductivity stainless steel, and a modified entrance seal adapted to fit the same opening which was employed for the glass beaker. The coolant chamber is within the shell of the thermal shield and is suspended from the top plate by the .006 inch stainless steel tube. The stainless steel tube, by virtue of its low conductivity acts effectively as an insulating agent to prevent a too rapid evaporation of liquid helium. The thermal web is suspended from the bottom of the coolant chamber and is fitted with two resistance heaters, thereby affording an annealing cycle while the specimen remains within the reduced atmosphere of the cylinder. The



substrate holder is affixed directly to the thermal web and merely holds the substrate in firm contact with the temperature cycling devices. This device will allow a much wider range than has heretofore been available for studies of stress, crystal imperfections and electro magnetic behavior of metal films. This system has not been constructed though it has been designed and the required material ordered.

In situ study of the thin film behavior will be confined, with the equipment designed, to collection of data as to the electrical resistivity of the films for comparison with the theoretical calculations by Fuchs [5] and Sondheimer [6] and the modifications proposed by Crittenden and Hoffman [2]. A much wider range of data may be collected by utilizing the feature of sealing the film in glass, as this will allow the specimen to be removed from the system for a great variety of measurements. Examples of the type data available are determination of stress induced within the film from measurements of curvature of the substrate, and nucleation centers as functions of substrate temperature at deposition.



CHAPTER III

ELECTRON BOMBARDMENT CRYOSTAT

The purpose in designing this equipment is to produce a known type of imperfection and observe its effects on the physical properties of the metal under controlled conditions wherein the entire history of the specimen will be fully known.

Imperfections in general may be produced in crystalline structures by cold work, condensation of material on a cold surface, bombardment with heavy particles, and bombardment with light charged particles. Of these methods of producing imperfections, only the bombardment with light particles appears to meet the requirement of producing a known type of imperfection. The other methods, e.g., cold work, produce several types of imperfections such as edge dislocations, ring dislocations, vacancies and interstitials. The idea here is to obtain a definite imperfection and follow its history. It is generally agreed that if copper is bombarded with electrons of one or two Mev energy the only imperfection expected is the "vacancy-interstitial" type. Once these are obtained, provisions must be made to hold the imperfection and also later to remove them. In addition there must be some means of observing the effects on the physical properties of the metal before the imperfections are introduced, during the growth of imperfection density, and during the removal or transition of the imperfections.

Theoretical considerations by Dienes [7] show that an increase in shear modulus by about 5 percent can be expected for 1 percent density of interstitials and a decrease of about 1 percent in shear modulus for 1 percent density of vacancies.



Preliminary experimental work along these lines has been conducted by Crittenden [8] and Dieckamp [9]. In this investigation there are portions of the history of each specimen after bombardment with electrons that are not fully known. This equipment is designed to eliminate this doubt.

To accomplish the above it was decided to use the Van de Graaff accelerator as a source of two Mev electrons in conjunction with a specially designed non-magnetic cryostat. The equipment consists of the following parts: dewar flask, nitrogen bucket, torsion pendulum, electron deflector and carriage.

A heavy outside brass vessel with a thin stainless steel liner form the dewar flask (see Figures 6, 7, 8). This flask had to meet two criteria; first it must be non-magnetic to facilitate future experiments where the applied magnetic field must be known and second, be of such a design as to allow the entrance of electrons of sufficient energy to displace the copper atoms to create the vacancies and interstitials. The non-magnetic design specification was met by the use of non-ferrous materials and 302 stainless steel. The scheme to allow the beam of two Mev electrons to reach the copper specimen located roughly in the center of the flask was solved by connecting the vacuum of the Van de Graaff directly into the side of the brass vessel. This way the electrons have to penetrate only the 12 mil stainless steel inside wall of the dewar flask to reach the specimen. In penetrating the wall the 2 Mev electrons lose approximately $1/4$ of their energy; this is allowable since 1 Mev electrons can produce the desired results.

From the standpoint of a dewar flask this design seems to be reasonable, that is, if material inside the flask is at liquid nitrogen



temperature -196°C , and 16 cm from the top, approximately 78,000 calories every 12 hours will leak into the flask by conduction down the inside walls. This amount of heat will vaporize a little over 2 liters in 12 hours which is not objectionable since the nitrogen bucket was designed to carry about 10 liters. It is possible to hold a specimen at -196°C for extended periods of time with only one refill of liquid nitrogen per day.

An inside wall thickness of .012 inch and 8 inch diameter will withstand a pressure of 75 psi before yielding, giving a safety factor of approximately five. It was expected that in designing this flask the pressures obtainable would go as low as 1×10^{-6} mm of mercury. This pressure was not reached; however, 1×10^{-5} mm of mercury (Hg) was reached and with a better diffusion pump 1×10^{-6} or even lower could probably be reached.

The nitrogen bucket is a double wall container whose function is to surround the copper specimen with a wall of liquid nitrogen (see Figures 9, 10). Here again provision had to be made to allow the entrance of the electrons. This was done simply by connecting the outer and inner walls by a 2 inch diameter tube which matched the Van de Graaff accelerator drift tube entrance on the dewar flask. This essentially forms a cylindrical wall of liquid nitrogen of approximately 8 inch outside diameter, 4 inch inside diameter and 18 inches high with a 2 inch tunnel through the side. These dimensions give a volume of liquid nitrogen of approximately 10 liters.

The torsion pendulum was designed and previously used by E.C. Crittenden, Jr. and H. Dieckamp [10] in the study of imperfections in copper introduced by cold work. This device consists of torsion pendulum



and furnace (see Figures 9, 12). The wire specimen which acts as the torsion element supports an inertial element of such a mass as not to cold work the copper wire while in oscillation. The inertial element is set into oscillation by an attached bar magnet which in turn is influenced by an externally applied magnetic field. By using a mirror at 45° and a concave mirror attached to the inertial element a light beam is directed from the top and reflected back out the top to indicate the motion of the pendulum. From the frequency of this motion and a knowledge of the dimensions of the wire, a true value of the shear modulus can be obtained.

The modifications made here were to arrange elements to fit into the non-magnetic dewar flask and nitrogen bucket, also to allow the furnace to be raised and lowered from outside the flask. In previous operations the furnace which is a ceramic resistor was lowered into position around the wire specimen. A rubber tube connected from the top of the furnace to the outside completed the quenching circuit which would allow the vaporizing nitrogen to flow along the wire specimen for rapid cooling. The torsion pendulum was then lowered into a cryostat and the furnace was essentially fixed since the rubber tube was frozen. A movable furnace was made by replacing the rubber tube with a stainless steel tube. Under the present plan of operation the specimen is to be mounted in the pendulum with furnace raised and placed in the dewar flask to obtain the frequency of oscillation before bombardment with electrons. During the bombardment, frequency determinations are made to indicate the imperfection density and once sufficient vacancies and interstitials are obtained bombardment is to be stopped. The furnace



is lowered into position around the wire specimen for thermal annealing. By closing the quenching vent the flow of nitrogen through the furnace is stopped and the annealing temperature can be raised to the desired level. This way the entire history of the wire specimen is known and controlled.

In view of the large amounts of energy received by the specimen during bombardment, it was felt that a special system is needed to keep the specimen temperature in the region of -196°C . This was solved by cycling the gas through a copper tube immersed in liquid nitrogen then into the dewar flask through the nitrogen bucket to be jetted onto the 10 mil wire (see Figure 9). Helium is used here rather than nitrogen since helium at this temperature has a specific heat of approximately five times that of nitrogen. A system was first devised where the helium could be recycled to save on the cooling liquid nitrogen. This was found to offer no advantage since the cold gas after leaving the recycling pump was approximately at room temperature.

The 10 mil copper wire specimens to be irradiated are approximately 1.5 inches in length and the focused electron beam is approximately a millimeter in diameter. Provisions had to be made for uniform irradiation of the specimen. From the dimensions involved it was found that a magnetic field of about a hundred gauss cm was sufficient to deflect the electrons enough to cover the entire length of the specimen. A complicated electron circuit is necessary to obtain a linear electron sweep. This was abandoned for an exponential sweep which gives uniform distribution over a complete cycle, that is, at any point the sum of the rise slope and the decay slope is equal to the sum of the slopes at any other



point. This type sweep was accomplished by applying a square voltage wave across the electro magnet spanning the drift tube of the accelerator (see Figure 13).

In replacing the retractable target of the target extension assembly of the Van de Graaff accelerator with the dewar flask, it was necessary to build a carriage to support the focusing magnet, drift tube and dewar flask. In the vicinity of the dewar flask the carriage was made of non-magnetic material (see Figure 8).



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APPENDIX I



FIGURE 1

Assembled View of Complete
Metal Film Evaporating Unit

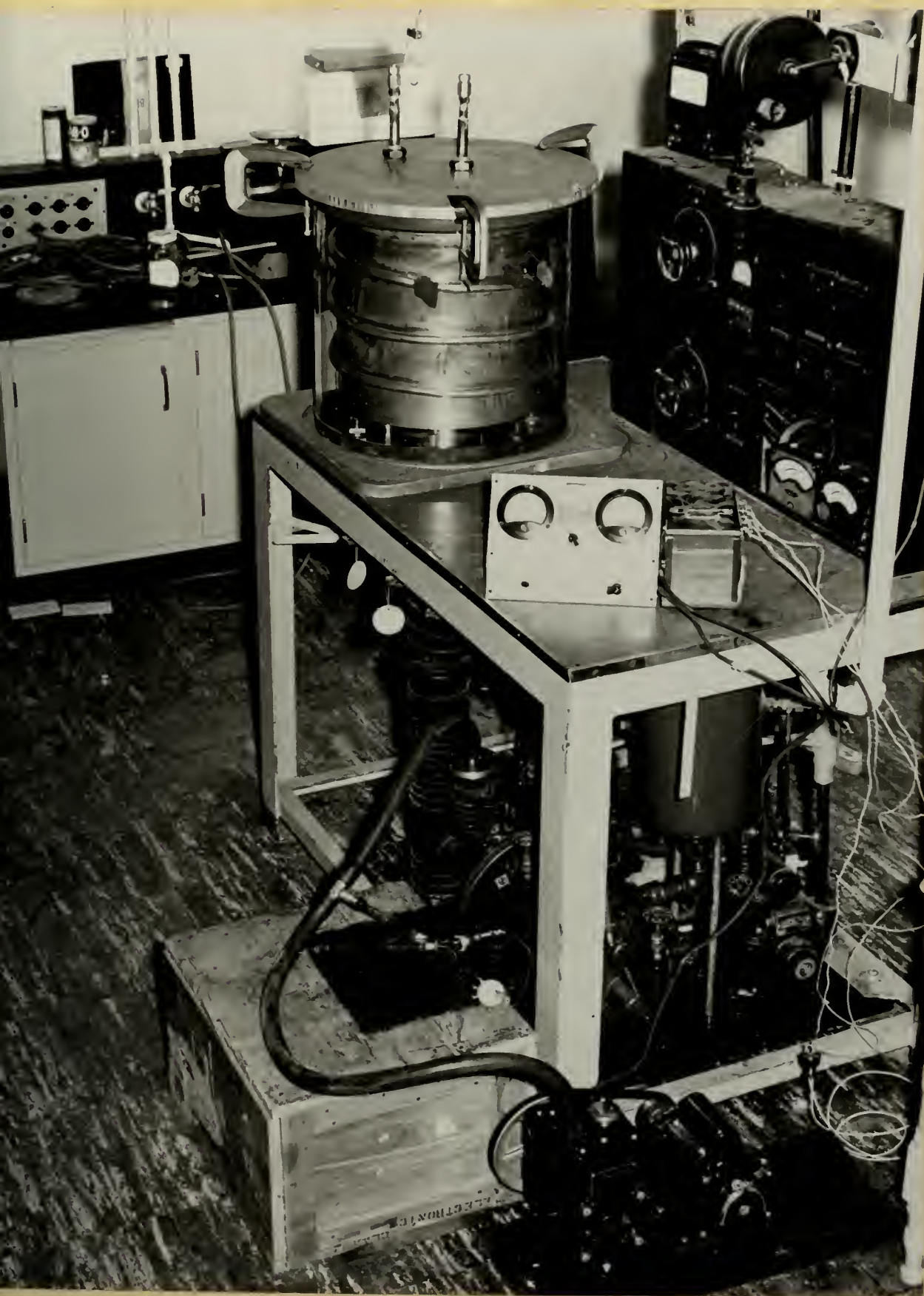


FIG 1



FIGURE 2

Testing Vacuum Developed
by MK 5 Pumping Units

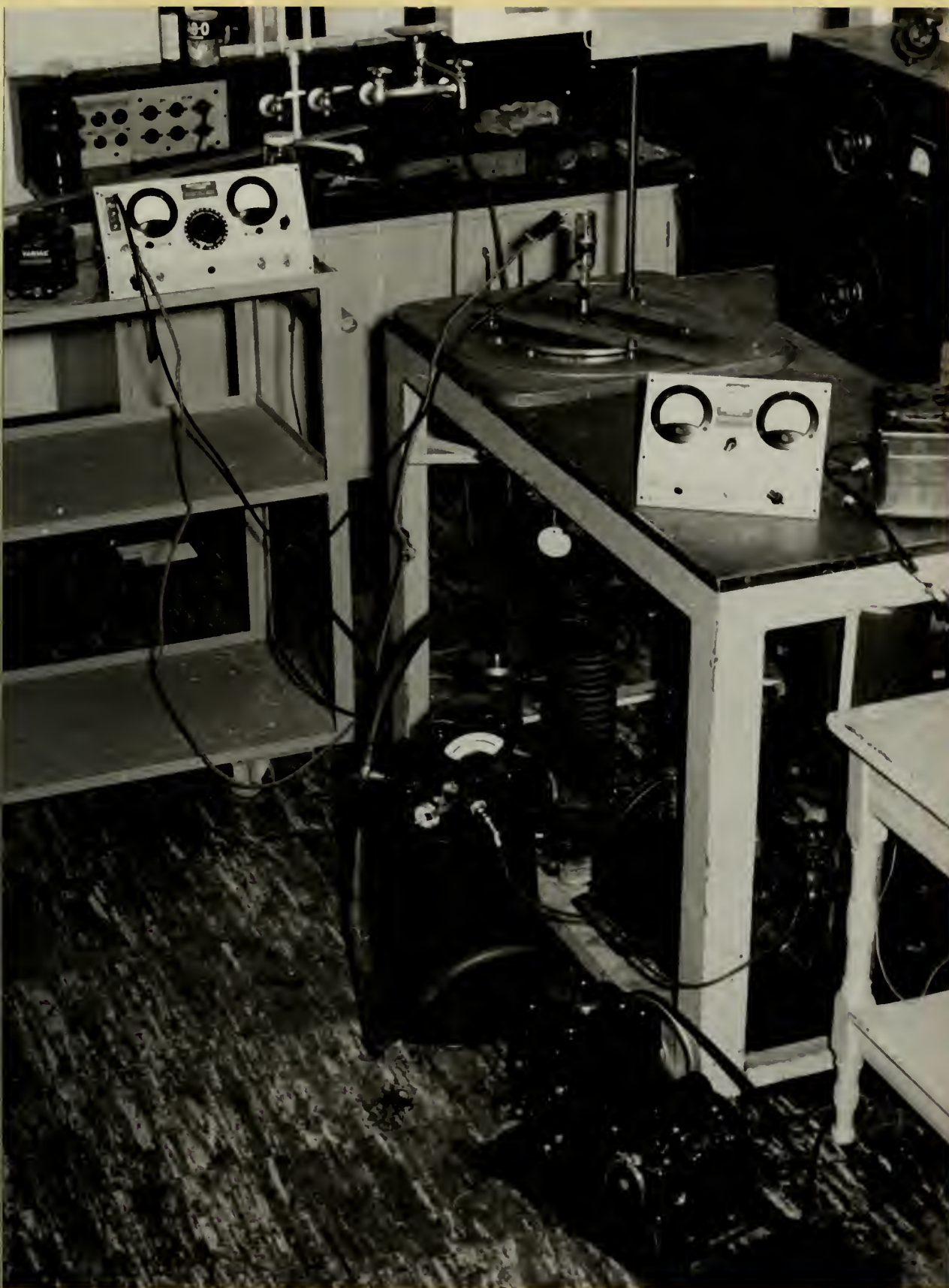


FIG 2



FIGURE 3

Exploded View of Component Parts Within the Glass Cylinder, Showing Thermalshield Suspended from the Top Plate and the Furnace and Charcoal Pump Installed on the Base Plate



FIG 3



FIGURE 4

Cross Section at Special Entrance Seal for Copper Tubing

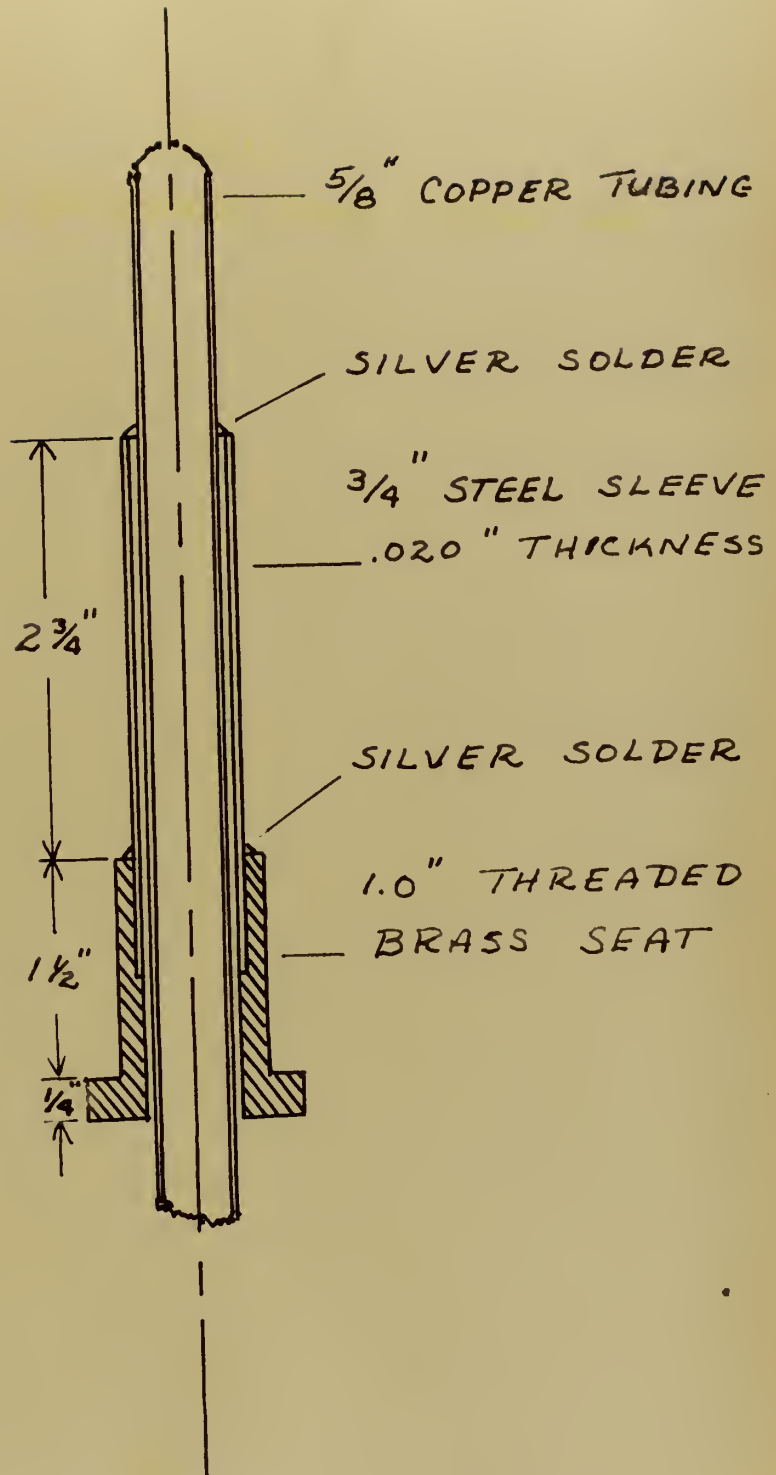


FIG 4



FIGURE 5

Cross Section of Liquid Helium Cooled Substrate Holder

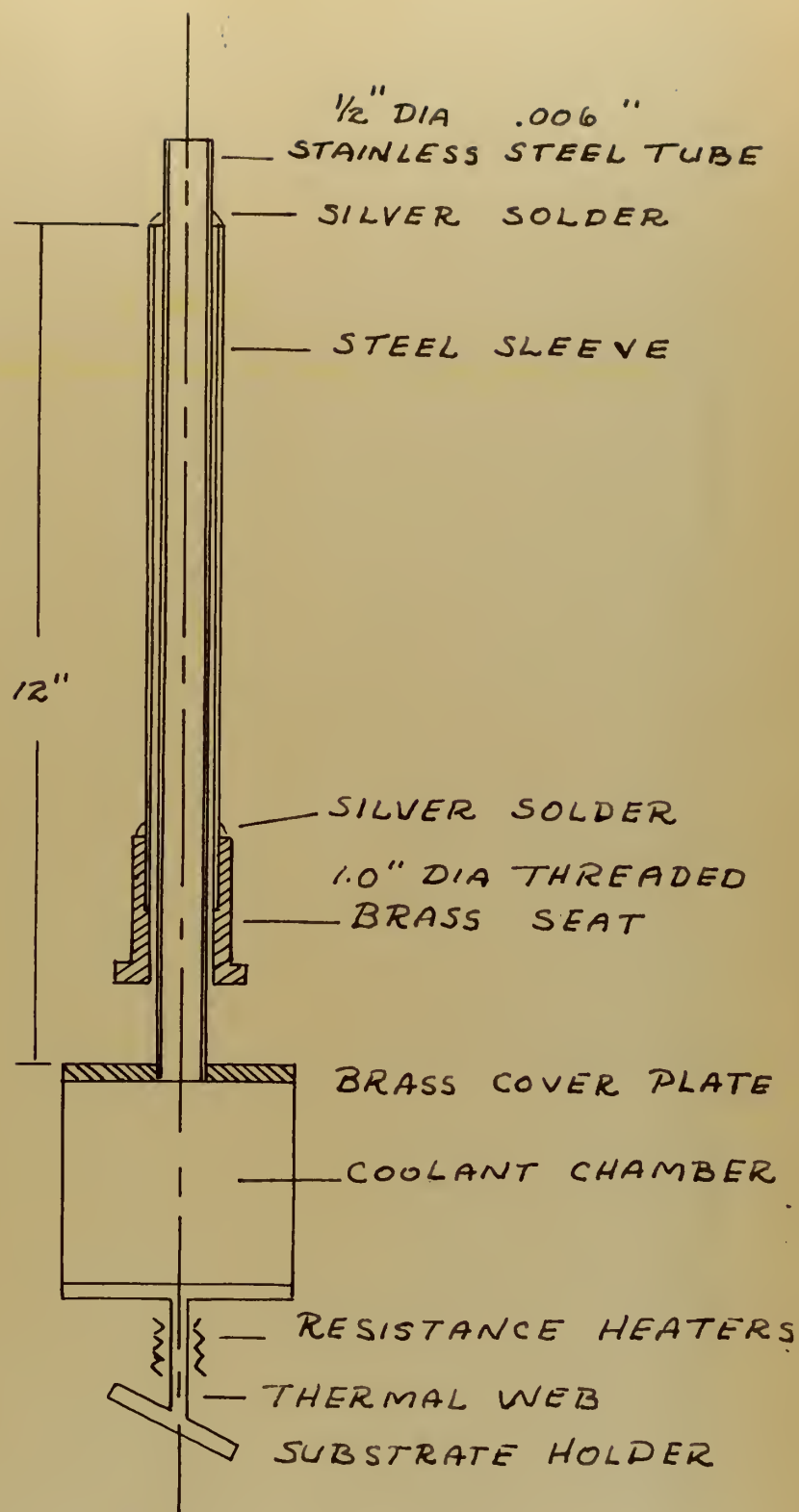


FIG 5



FIGURE 6

Arrangement Used to Evacuate the Outside Brass Vessel of the Dewar Flask

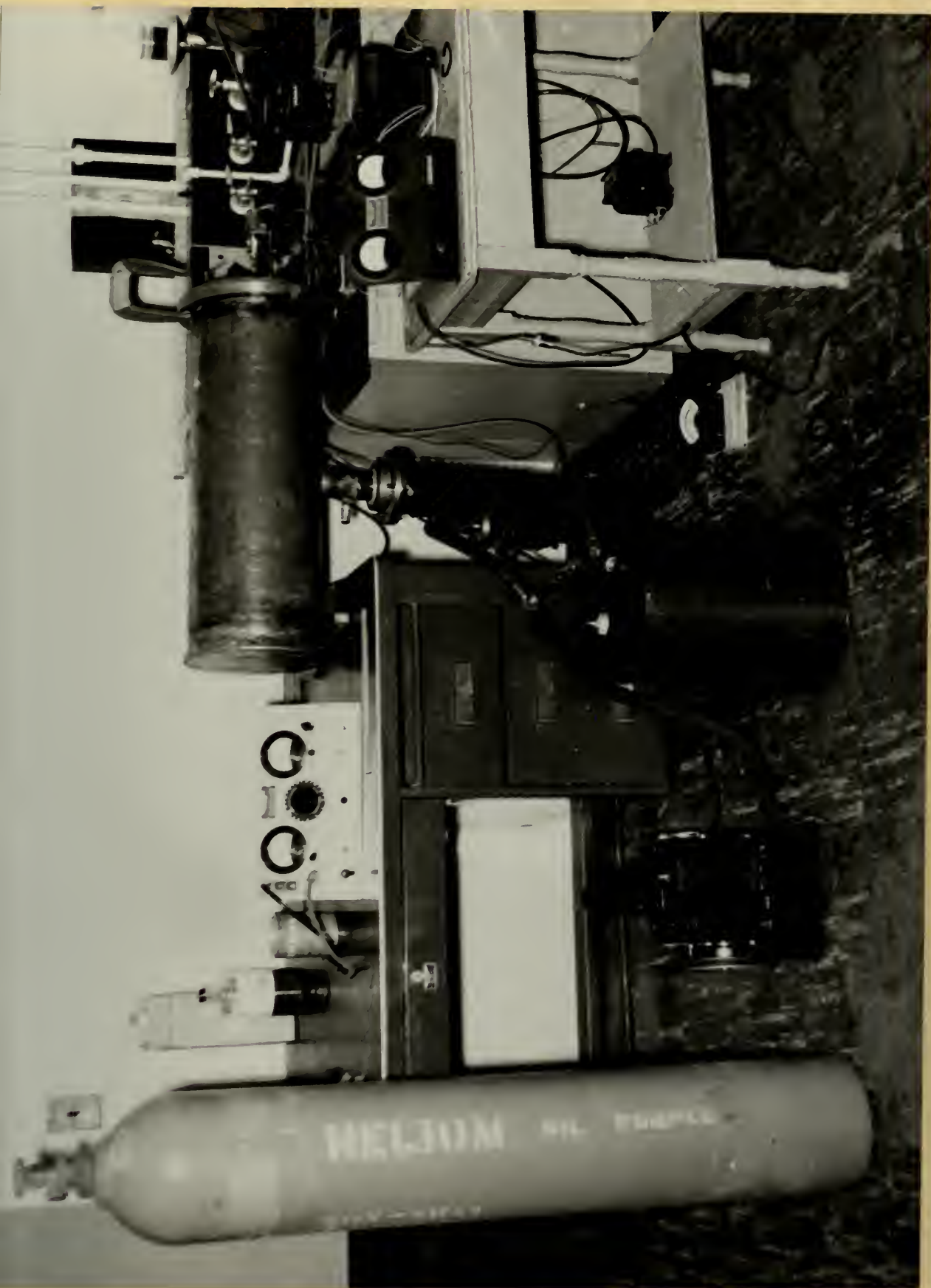


FIG 6



FIGURE 7

Complete Dewar Flask Being Evacuated and Checked for Leaks

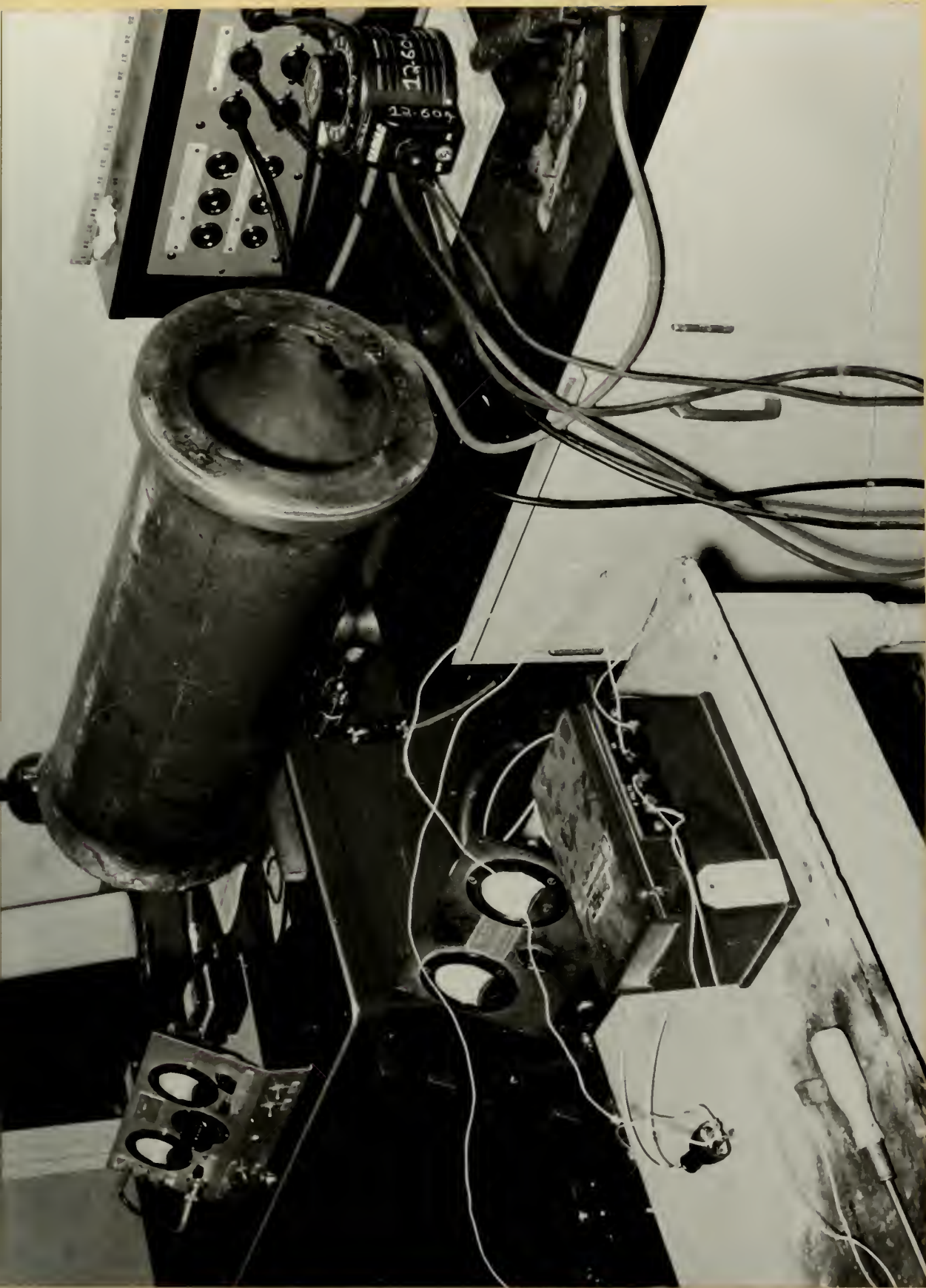


FIG 7



FIGURE 8

Drift Tube, Focusing Magnet and Dewar Flask Mounted on Special Carriage



FIG 8

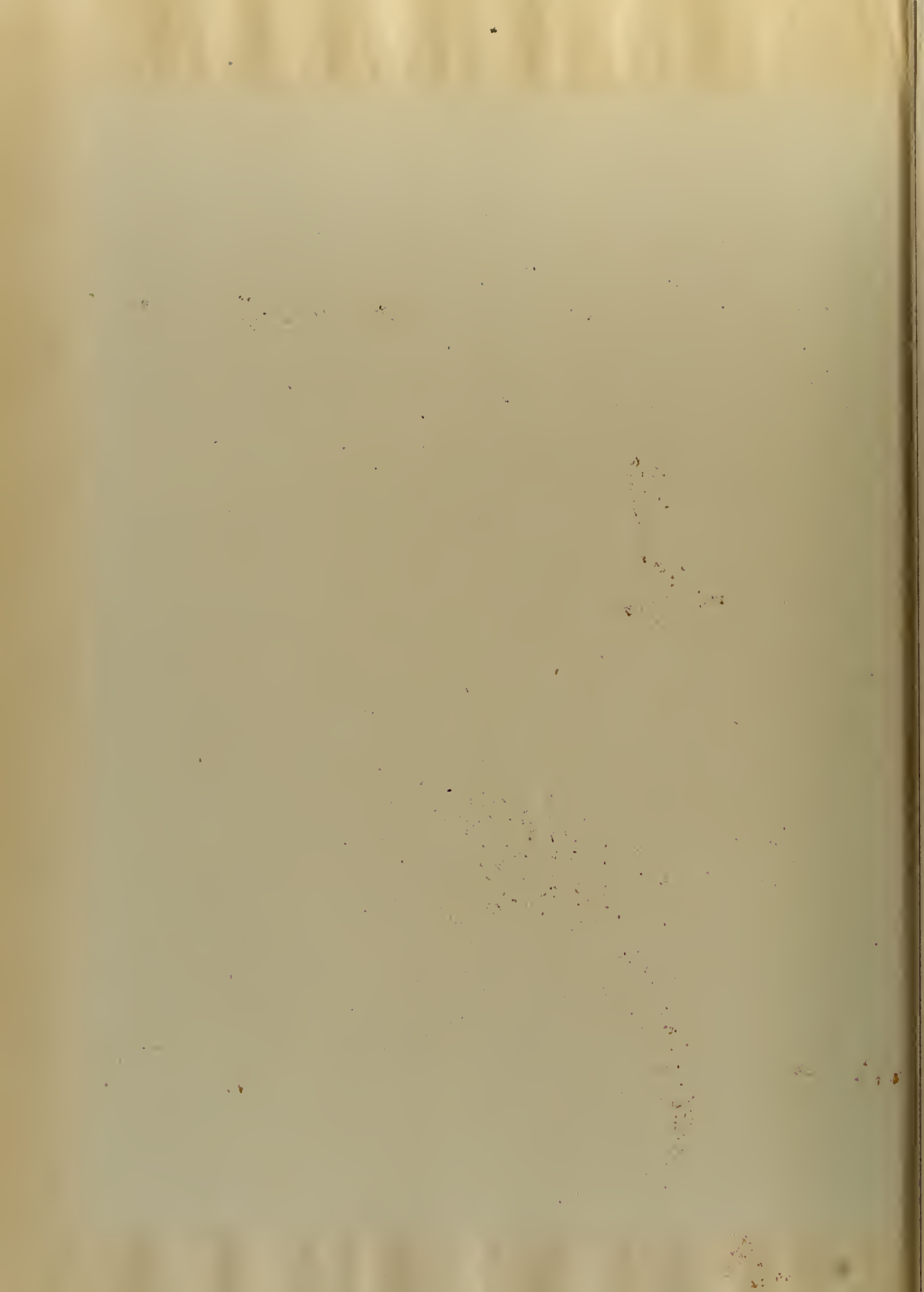


FIGURE 9

Nitrogen Bucket, Cooling Tube and Torsion Pendulum Disassembled

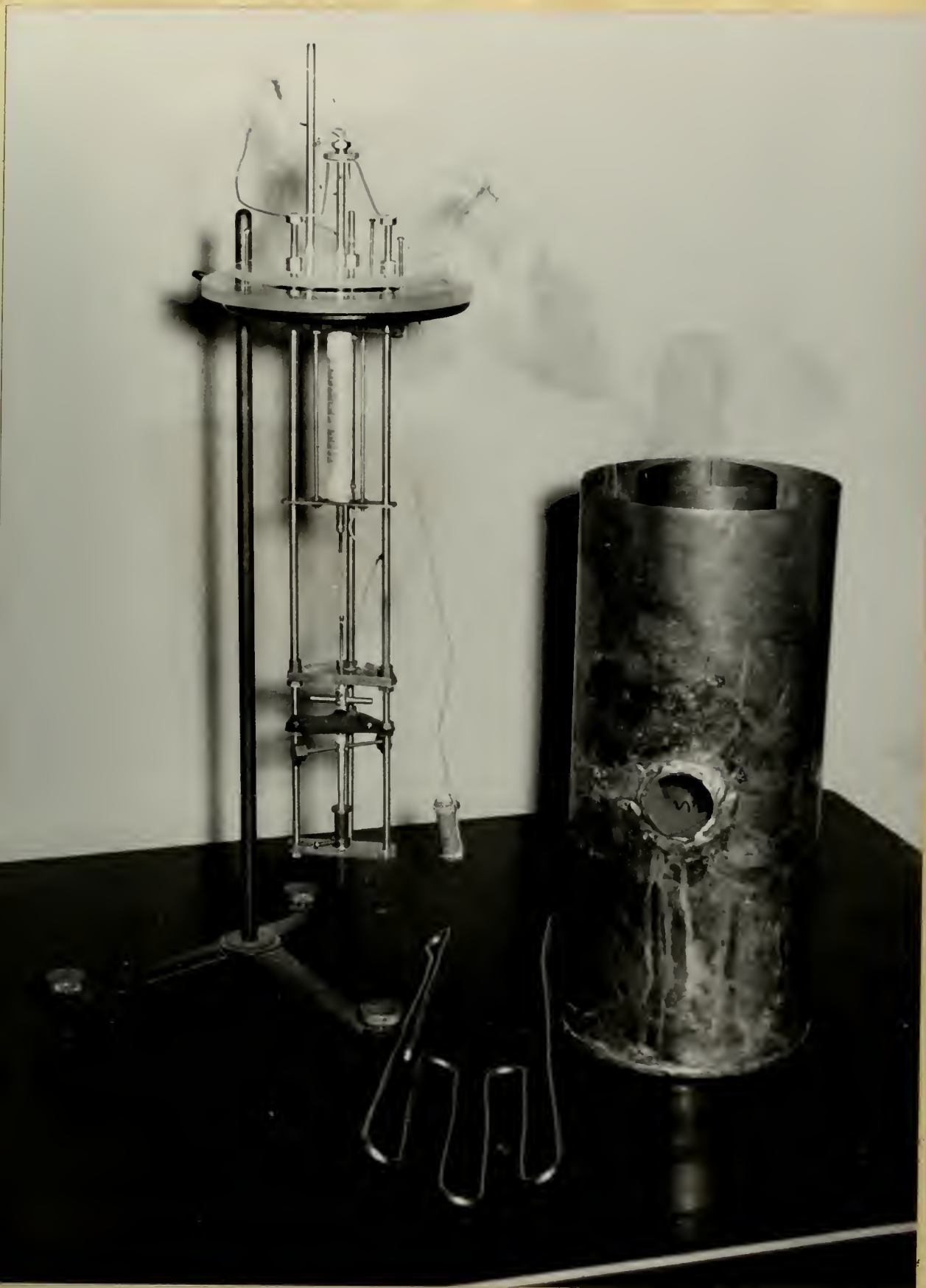


FIG 9



FIGURE 10

Nitrogen Bucket with Torsion Pendulum and Cooling Tube in Place

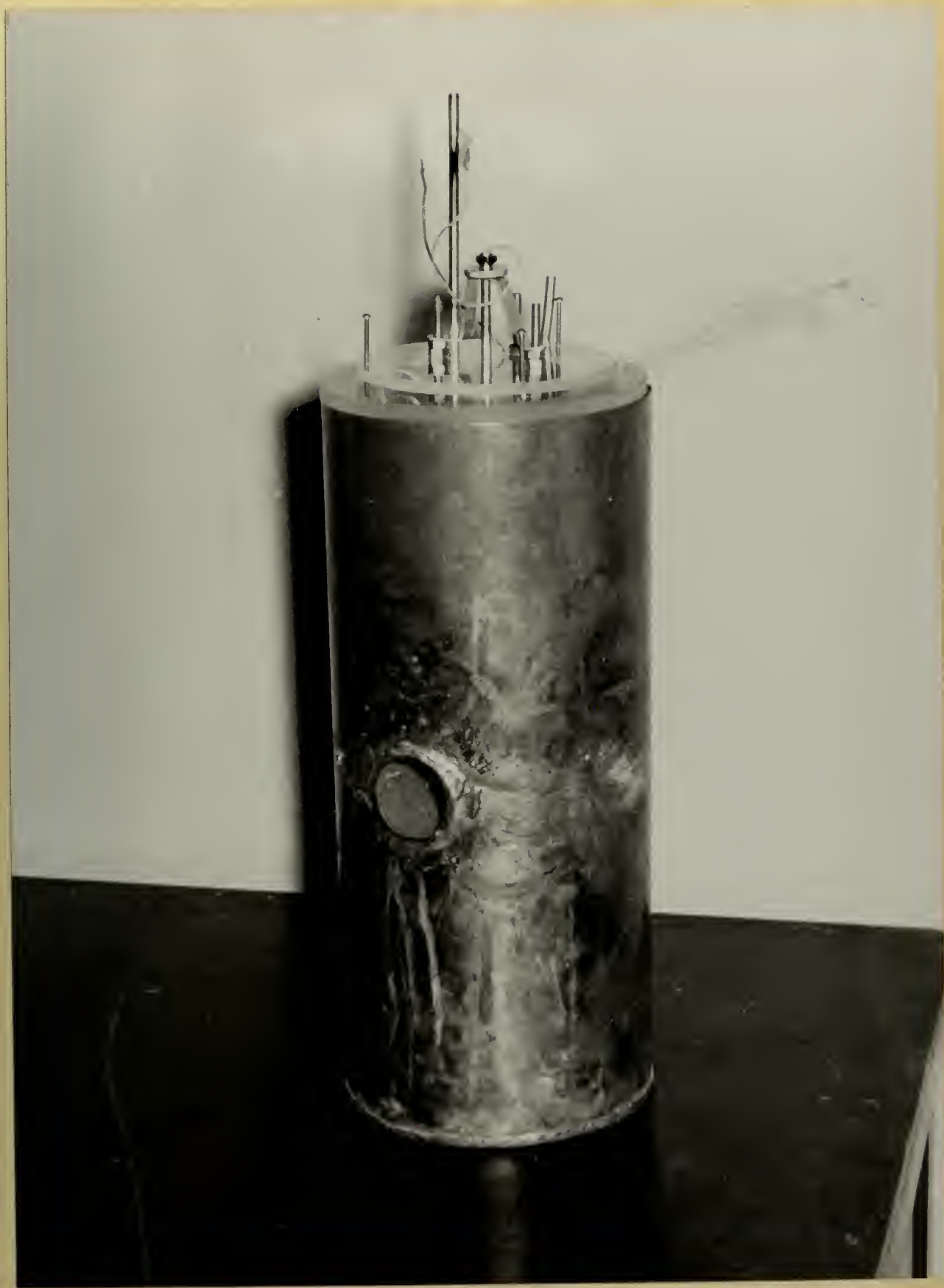


FIG 10



FIGURE 11

Dewar Flask Attached to Van de Graaff for Evacuation

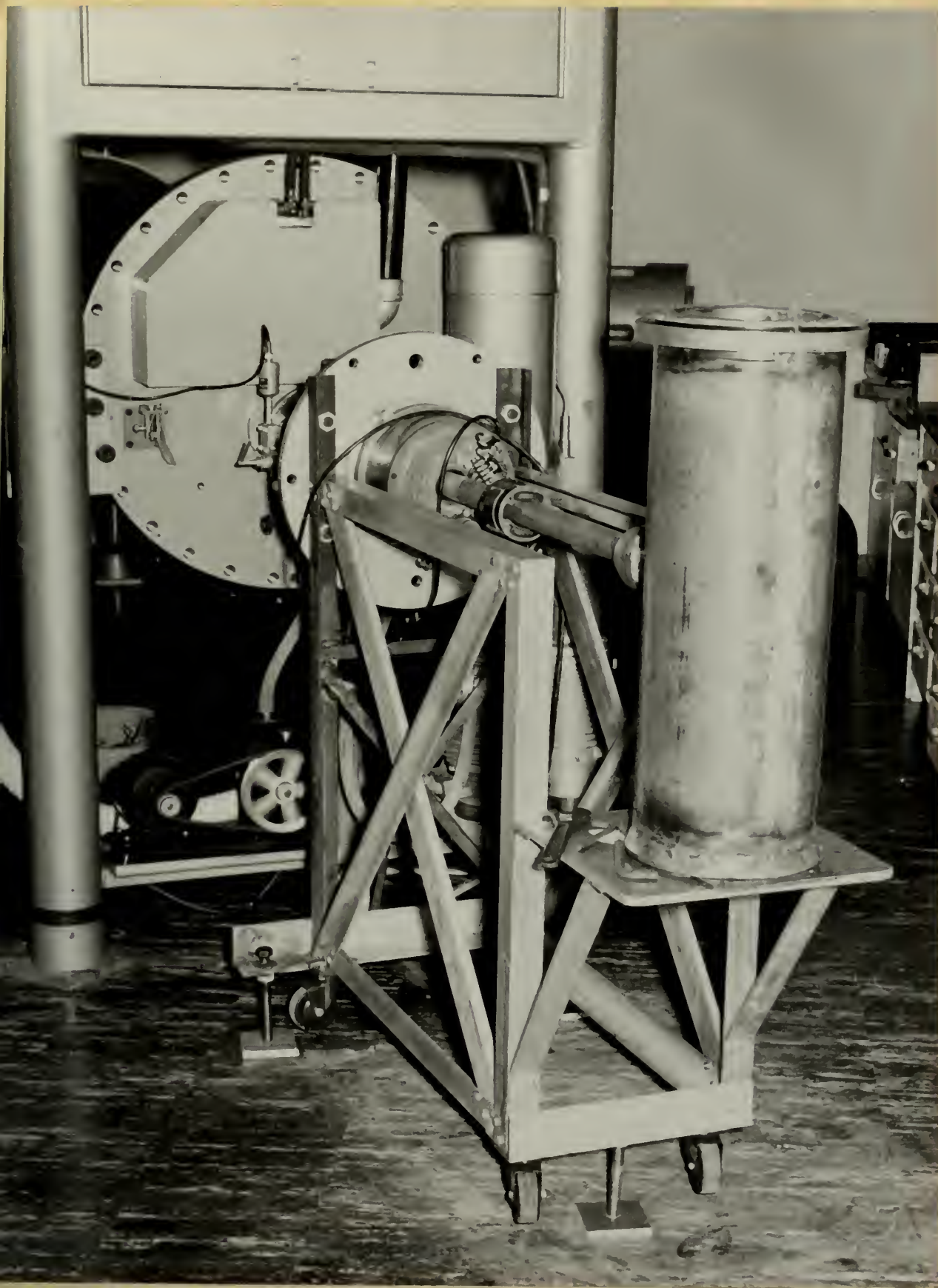


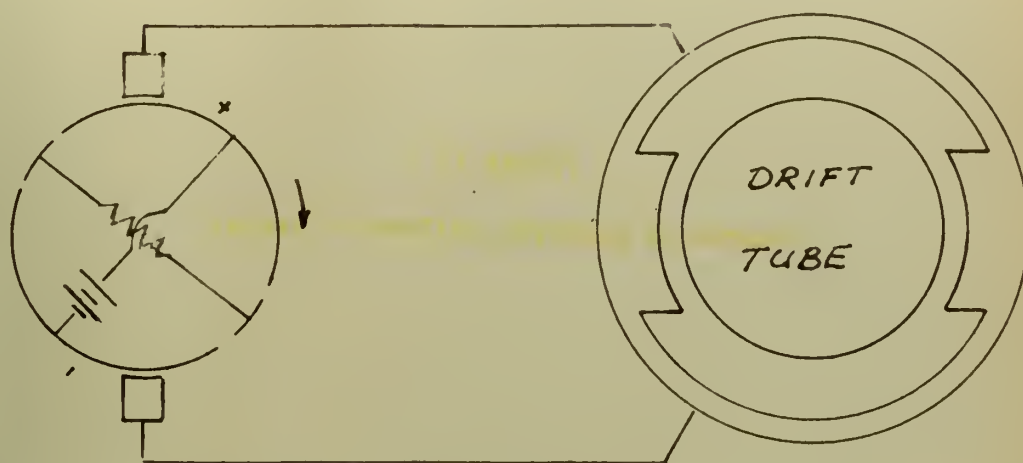
FIG 11





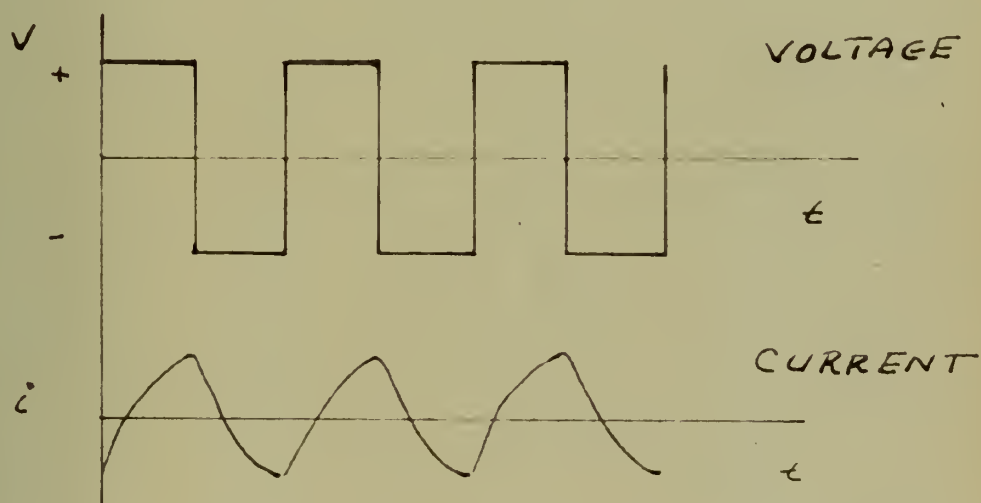
FIGURE 13

Diagram of Electron Deflector Circuit



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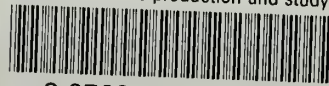
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